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PN=03-011003

# METHOD FOR CONTROLLING PESTS
[YUGAI SEIBUTSU NO BOJO HOHO]

Terumasa Komyoji, et al.

UNITED STATES PATENT AND TRADEMARK OFFICE
Washington, D.C. January 2004

Translated by: FLS, Inc.

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APPLICANT (71): ISHIHARA SANGYO KAISHA, LTD.

TITLE (54): METHOD FOR CONTROLLING PESTS

FOREIGN TITLE [54A]: YUGAI SEIBUTSU NO BOJO HOHO

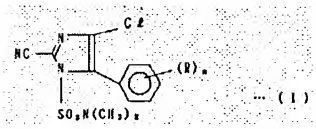
Specifications

1. Title of the Invention

Method for Controlling Pests

2. Claim(s)

A method for controlling pests characterized by applying, to certain places where pests break out or there is the possibility of an outbreak, an aqueous dispersion containing at least one kind of imidazole-based compound represented by the general formula (I) below and a sorbitan higher fatty acid ester surfactant.



(Where **R** in the formula is a halogen atom, an alkyl group, or an alkoxyl group, and **n** is an integer from 0 to 5.)

3. Detailed Specifications

(Field of Industrial Application)

The present invention relates to a method for controlling pests by applying an aqueous solution containing a specific imidazole-based compound and a sorbitan higher fatty acid ester surfactant.

(Prior Art)

Specific imidazole-based compounds which are useful as pest control agents are described in European Patent Application No. 298,196. Although these compounds which are contained and applied with a sorbitan higher fatty acid ester surfactant are generally described here, they are not specifically disclosed.

(Problems to Be Solved by the Invention)

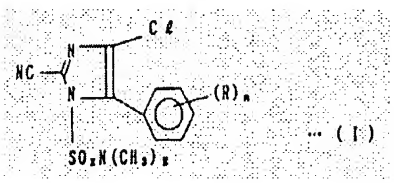
During actual application of a pest control agent, on the other hand, the control of different types of pests causing various diseases and having different periods of outbreak as completely as possible, and at the same time, consideration of a reduction in control costs and its effect on contaminating the environment, and reducing the amount of the active ingredient compound used are required.

(Means for Solving the Problems)

The inventors of the present invention discovered that the pest controlling effect could be improved remarkably and the amount of the active ingredient compound used could be reduced as compared to a compound without any sorbitan higher fatty acid ester surfactant by containing and applying the aforementioned imidazole-based compound together with the sorbitan higher fatty acid ester surfactant, and they accomplished the present invention.

(Disclosure of the Invention)

The present invention relates to method for controlling pests characterized by applying, to certain places where pests break out or there is the possibility of an outbreak, an aqueous dispersion containing at least one kind of imidazole-based compound represented by the general formula (I) below and a sorbitan higher fatty acid ester surfactant as the active ingredient compound.



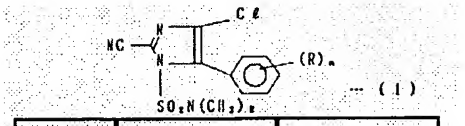
... (I)

(Where **R** in the formula is a halogen atom, an alkyl group, or an alkoxyl group, and **n** is an integer from 0 to 5.).

A fluorine, chlorine, bromine or iodine atom can be cited for the halogen atom represented by **R** in the active ingredient compound of the general formula (I). A methyl group, ethyl group, propyl group, and the like can be cited as examples of the alkyl constituent having 1 to 6 carbons of the alkyl group and alkoxyl group represented by **R**. These groups include those with the structural isomerism of a straight chain or branched fatty chain. Moreover, when **n** is 2 or greater, **R** may be homogeneous or heterogeneous.

The active ingredient compound represented by the aforesaid general formula (I) is a known compound described in European Patent Application No. 298,196, and typical examples thereof are shown in Table 1.

Table 1



... ( I )

| Compound No. | (R)n | Physical properties (melting point °C) |
|---|---|---|
| 1 | Unsubstituted | 109 ~ 112 |
| 2 | 4 - $CH_3$ | 133 ~ 134 |
| 3 | 3 - $CH_3$ | - |
| 4 | 2 - $CH_3$ | 93 ~ 96 |
| 5 | 3,4-$(CH_3)_2$ | - |
| 6 | 4 - $OCH_3$ | - |
| 7 | 4 - Cℓ | 117 ~ 120 |
| 8 | 2 - Cℓ | 113 ~ 117 |
| 9 | 3,4-$Cℓ_2$ | - |
| 10 | 4 - F | 120 ~ 122 |
| 11 | 4 - Br | - |
| 12 | 4 - $C_3H_7$(i) | 135 ~ 138 |
| 13 | 4 - $C_3H_8$ | 110 ~ 112 |
| 14 | 3 - Cℓ | 96 ~ 99 |
| 15 | 3 - F | - |
| 16 | 2 - F | - |

Meanwhile, the sorbitan higher fatty acid ester surfactant used in the method of the present invention should be a surfactant to which a hydrophilic group and lipophilic group has been added to dehydrated and cyclized sorbitols or a surfactant containing them as the active ingredient. For example, Sorgen TW-20, TW-60 and TW-80 (trade names, all available from Daiichi Pharmaceutical Co., Ltd.), Pionin D-941, D-944, D-945 and D-945T (trade names, all available from Takemoto Oil & Fat Co., Ltd.), Sylvan T-20, T-60 and T-80 (trade names, all available from Matsumoto Yushi-Seiyaku Co., Ltd), Sorbon T-20, T-40, T-60, T-65, T-80 and T-85 (trade names, all available from Toho Chemical Industry Co., Ltd.), and Reodole TW-L106, TW-L120, TW-P120, TW-S106, TW-S120, TW-S320, TW-O106, TW-0120 and TW-O320, Emazol L-120, P-120, S-120, O-120 and O-105, and Approach [transliteration] BI (trade names all available from Kao Corp.),

and the like may be used. Of these surfactants, Approach [transliteration] BI is especially desirable.

The term "pest" in the present invention refers to an agricultural or horticultural pest, such as vegetable pathogens which cause diseases, such as rice blast, rice sheath blight, anthracnose of cucumber, cucumber powdery mildew and downy mildew, leafspot and blight of tomato, tomato ring spot disease, black spot of citrus, filamentous fungi of citrus, Japanese pear scab fungus, Alternaria leaf spot on apple, downy mildew of grape, various Botrytis cinerea Persoon, cottony rot, and rust; vegetable pathogens causing soil blight, such as the genera *Fusarium*, *Pythium*, *Rhizoctonia*, *Verticillium*, and *Plasmodiophora*; insects, such as leafhopper, diamondback moth, green rice leafhopper, adzuki bean beetle, common cutworm, and green peach aphid; mites, such as two-spotted spider mite, carmine spider mite, and citrus red mite; nematodes, such as sweet potato root-knot nematode; and the like are cited as examples.

The stems and leaves of useful plants, the soil, and the like are cited as the certain places where pests break out or there is the possibility of an outbreak, but most cases of outbreak involve the stems and leaves of useful plants.

Aqueous dispersions in which the aforesaid imidazole-based compound, which has been preformulated as a wettable powder, flowable agent, emulsion, or the like, has been dispersed in water and a sorbitan fatty acid ester surfactant has been added to this; an aqueous dispersion in which the aforesaid imidazole-based compound and the sorbitan higher fatty acid ester surfactant, which have been premixed and formulated, are dispersed

in water; or an aqueous dispersion in which the imidazole-based compound and sorbitan fatty acid ester surfactant are dispersed in water in a corresponding method is cited as the aqueous dispersion used in the method of the present invention. The amount of the sorbitan higher fatty acid ester surfactant blended in the aqueous dispersion is normally 0.01 to 50 mL (or g), and desirably, 0.1 to 5 mL (or g) per liter of the aforesaid aqueous dispersion. Since the concentration of the aforesaid imidazole-based compound used as an active ingredient compound in the method of the present invention varies depending on the conditions of formulating the chemical, the period of application, the type of target pest, and the like, and is not generally specified, it is 0.1 to 10,000 ppm, and desirably, 1 to 2,000 ppm in the case of leaf/stem treatment, and 10 to 100,000 g/ha, and desirably, 200 to 20,000 g/ha in the case of a soil treatment.

Moreover, in the method of the present invention, other agricultural chemicals, such as bactericides, pesticides, miticides, nematicides, antiviral agents, attractants, plant growth regulators, and the like, can be mixed with the imidazole-based compound. In this case, far more superior effects can be exhibited. In particular, typical examples of the other agrochemicals (common names) include azole-based compounds, such as tritriflumizole; quinoxaline-based compounds, such as quinomethionate; dithiocarbamate-based compounds, such as Anzeb; organochlorine-based compounds, such as chlorothalonil; benzimidazole-based compounds, such as benomyl; pyridinamine-compounds, such as fluazinam; dianoacetamide-based compounds, such as cymoxanil;

phenylamide-based compounds, such as metaraxyl and oxadixyl; sulfenic acid-based compounds, such as dichlofluanide; copper-based compounds, such as ferric hydroxide; isoxazole-based compounds, such as hydroxyisoxazole; organophosphorus-based compounds, such as phosethyl aluminum; dicarboxyimide-based compounds, such as procymidone; benzanilide-based compounds, such as flutolanil; benzamide-based compounds, such as (RS)-4-chloro-N-(cyano(ethoxy)methyl)benzamide, etc.

In order to describe the method of the present invention in more detail, test examples are cited next but the present invention is not limited to just these test examples.

Test Example 1: Test on effect of treating Phytophthora infestans (production of aqueous dispersion)

1. Active ingredient compound — 20 parts by weight
   (any of the compounds in aforesaid Table 1)

2. Zeeklite (kaolin) — 62.4 parts by weight
   (trade name, available from Zeeklite Ind.)

3. Carplex #80 — 12.0 parts by weight
   (trade name, fine amorphous silicon dioxide particles available from Shionogi & Co., Ltd.)

4. Sorpol 5073 — 2.4 parts by weight
   (trade name, polyoxyethylene alkylaryl ether sulfate; available from Toho Chemical Industry Co., Ltd.)

5. Sorbol 5060 — 1.6 parts by weight
   (trade name, alkylaryl sulfonate; available from Toho Chemical Industry Co., Ltd.)

6. Lavelin FA-N — 1.6 parts by weight
   (trade name for sodium naphthalene sulfonate-formalin condensate; available from Daiichi Pharmaceutical Co., Ltd.)

A wettable powder mixture of each of the above ingredients was dispersed in water so that the active ingredient was diluted to the prescribed

concentration and 1 mL/L of Approach [as transliterated] BI (trade name, available from Kao Corp.) was added next to prepare an aqueous dispersion.

Moreover, except that no Approach [as transliterated] BI was added, each aqueous solution sample was prepared in the same manner to obtain the respective comparison products.

(Biological Test Method and Results)

Tomato (cultivar: Ponterosa) was cultivated in 7.5 cm plastic pots, and inoculated by being sprayed with a suspension of pathogenic fungus zoospores when the seedlings reached the 4-leaf stage. After 6 hours, the plants were sprayed with 10 mL of each aqueous dispersion sample using a spray can. After keeping them at 22 to 24°C for 5 days in a constant temperature room, the area of the disease spots was examined, the control index was found according to the evaluation standards given below, and the results in Table 2 were obtained.

Evaluation Criteria

The extent of an outbreak on the plant samples was observed visually during the examination and a 7-level control index was obtained as follows for the control effect.

(Control Index) (Extent of Outbreak)

7: No disease spots confirmed at all
6: Area of disease spots less than 5% of untreated section
5: Area of disease spots less than 5 to 10% of untreated section
4. Area of disease spots less than 10 to 25% of untreated section
3. Area of disease spots less than 25 to 40% of untreated section
2. Area of disease spots less than 40 to 70% of untreated section
1. Area of disease spots 70% or more of untreated section

Table 2

| Aqueous dispersion sample | Active ingredient compound | Active ingredient concentration (ppm) | Control index |
|---|---|---|---|
| Aqueous dispersion No. 1 | Compound No. 1 | 31 | 7 |
| Comparison product of aqueous dispersion No. 1 | " | " | 2 |
| Aqueous dispersion No. 2 | Compound No. 8 | 31 | 7 |
| Comparison product of aqueous dispersion No. 2 | " | " | 1 |
| Aqueous dispersion No. 3 | Compound No. 14 | 31 | 5 |
| Comparison product of aqueous dispersion No. 3 | " | " | 1 |

Test Example 2: Test of effect in treating cucumber downy mildew

(Preparation of aqueous dispersion)

Each aqueous dispersion sample was prepared as in Test Example 1.

((Biological Test Method and Results)

Cucumber (cultivar: Suiyo) was cultivated in 7.5 cm plastic pots and inoculated by being sprayed with a suspension of *Pseudoperonopora humuli* zoospores when the seedlings reached the 2-leaf stage. After 24 hours, each aqueous dispersion sample was sprayed at a proportion of 10 mL per 0.25$m^2$. After maintaining this at 22 to 24°C for 6 days in a constant temperature room, the area of the disease spots on the 1$^{st}$ leaf was examined, the control index was found according to the evaluation standards in aforesaid Test Example 1, and the results in Table 3 were obtained.

Table 3

| Aqueous dispersion sample | Active ingredient compound | Active ingredient concentration (ppm) | Control index |
|---|---|---|---|
| Aqueous dispersion No. 4 | Compound No. 1 | 2 | 7 |
| Comparison product of aqueous dispersion No. 4 | " | " | 3 |
| Aqueous dispersion No. 5 | Compound No. 8 | 31 | 7 |
| Comparison product of aqueous dispersion No. 5 | " | " | 2 |
| Aqueous dispersion No. 6 | Compound No. 12 | 31 | 5 |
| Comparison product of aqueous dispersion No. 6 | " | " | 1 |
| Aqueous dispersion No. 6 | Compound No. 14 | 8 | 7 |
| Comparison product of aqueous dispersion No. 6 | " | " | 1 |

(Advantages of the Invention)

According to the control method of the present invention, and particularly, the pest control effect and effect for treating plant diseases may be improved remarkably, which is effective in reducing the amount of active ingredient compound used.

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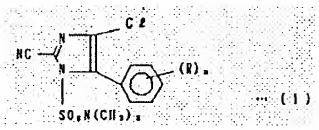

(Where **R** in the formula is a halogen atom, an alkyl group, or an alkoxyl group, and **n** is an integer from 0 to 5.)

3. Detailed Specifications

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The present invention relates to a method for controlling pests by applying an aqueous solution containing a specific imidazole-based compound and a sorbitan higher fatty acid ester surfactant.

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Specific imidazole-based compounds which are useful as pest control agents are described in European Patent Application No. 298,196. Although these compounds which are contained and applied with a sorbitan higher fatty acid ester surfactant are generally described here, they are not specifically disclosed.

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The inventors of the present invention discovered that the pest controlling effect could be improved remarkably and the amount of the active ingredient compound used could be reduced as compared to a compound without any sorbitan higher fatty acid ester surfactant by containing and applying the aforementioned imidazole-based compound together with the sorbitan higher fatty acid ester surfactant, and they accomplished the present invention.

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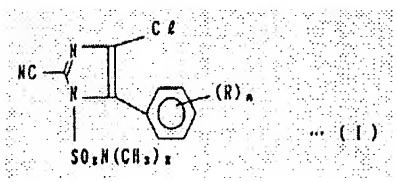

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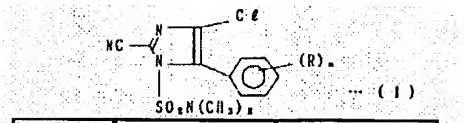


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| 12 | 4 - $C_3H_7$(i) | 135 ~ 138 |
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| 14 | 3 - Cℓ | 96 ~ 99 |
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Meanwhile, the sorbitan higher fatty acid ester surfactant used in the method of the present invention should be a surfactant to which a hydrophilic group and lipophilic group has been added to dehydrated and cyclized sorbitols or a surfactant containing them as the active ingredient. For example, Sorgen TW-20, TW-60 and TW-80 (trade names, all available from Daiichi Pharmaceutical Co., Ltd.), Pionin D-941, D-944, D-945 and D-945T (trade names, all available from Takemoto Oil & Fat Co., Ltd.), Sylvan T-20, T-60 and T-80 (trade names, all available from Matsumoto Yushi-Seiyaku Co., Ltd), Sorbon T-20, T-40, T-60, T-65, T-80 and T-85 (trade names, all available from Toho Chemical Industry Co., Ltd.), and Reodole TW-L106, TW-L120, TW-P120, TW-S106, TW-S120, TW-S320, TW-O106, TW-0120 and TW-O320, Emazol L-120, P-120, S-120, O-120 and O-105, and Approach [transliteration] BI (trade names all available from Kao Corp.),

and the like may be used. Of these surfactants, Approach [transliteration] BI is especially desirable.

The term "pest" in the present invention refers to an agricultural or horticultural pest, such as vegetable pathogens which cause diseases, such as rice blast, rice sheath blight, anthracnose of cucumber, cucumber powdery mildew and downy mildew, leafspot and blight of tomato, tomato ring spot disease, black spot of citrus, filamentous fungi of citrus, Japanese pear scab fungus, Alternaria leaf spot on apple, downy mildew of grape, various Botrytis cinerea Persoon, cottony rot, and rust; vegetable pathogens causing soil blight, such as the genera *Fusarium*, *Pythium*, *Rhizoctonia*, *Verticillium*, and *Plasmodiophora*; insects, such as leafhopper, diamondback moth, green rice leafhopper, adzuki bean beetle, common cutworm, and green peach aphid; mites, such as two-spotted spider mite, carmine spider mite, and citrus red mite; nematodes, such as sweet potato root-knot nematode; and the like are cited as examples.

The stems and leaves of useful plants, the soil, and the like are cited as the certain places where pests break out or there is the possibility of an outbreak, but most cases of outbreak involve the stems and leaves of useful plants.

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in water; or an aqueous dispersion in which the imidazole-based compound and sorbitan fatty acid ester surfactant are dispersed in water in a corresponding method is cited as the aqueous dispersion used in the method of the present invention. The amount of the sorbitan higher fatty acid ester surfactant blended in the aqueous dispersion is normally 0.01 to 50 mL (or g), and desirably, 0.1 to 5 mL (or g) per liter of the aforesaid aqueous dispersion. Since the concentration of the aforesaid imidazole-based compound used as an active ingredient compound in the method of the present invention varies depending on the conditions of formulating the chemical, the period of application, the type of target pest, and the like, and is not generally specified, it is 0.1 to 10,000 ppm, and desirably, 1 to 2,000 ppm in the case of leaf/stem treatment, and 10 to 100,000 g/ha, and desirably, 200 to 20,000 g/ha in the case of a soil treatment.

Moreover, in the method of the present invention, other agricultural chemicals, such as bactericides, pesticides, miticides, nematicides, antiviral agents, attractants, plant growth regulators, and the like, can be mixed with the imidazole-based compound. In this case, far more superior effects can be exhibited. In particular, typical examples of the other agrochemicals (common names) include azole-based compounds, such as tritriflumizole; quinoxaline-based compounds, such as quinomethionate; dithiocarbamate-based compounds, such as Anzeb; organochlorine-based compounds, such as chlorothalonil; benzimidazole-based compounds, such as benomyl; pyridinamine-compounds, such as fluazinam; dianoacetamide-based compounds, such as cymoxanil;

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In order to describe the method of the present invention in more detail, test examples are cited next but the present invention is not limited to just these test examples.

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4. Sorpol 5073 — 2.4 parts by weight
   (trade name, polyoxyethylene alkylaryl ether sulfate; available from Toho Chemical Industry Co., Ltd.)

5. Sorbol 5060 — 1.6 parts by weight
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A wettable powder mixture of each of the above ingredients was dispersed in water so that the active ingredient was diluted to the prescribed

concentration and 1 mL/L of Approach [as transliterated] BI (trade name, available from Kao Corp.) was added next to prepare an aqueous dispersion.

Moreover, except that no Approach [as transliterated] BI was added, each aqueous solution sample was prepared in the same manner to obtain the respective comparison products.

(Biological Test Method and Results)

Tomato (cultivar: Ponterosa) was cultivated in 7.5 cm plastic pots, and inoculated by being sprayed with a suspension of pathogenic fungus zoospores when the seedlings reached the 4-leaf stage. After 6 hours, the plants were sprayed with 10 mL of each aqueous dispersion sample using a spray can. After keeping them at 22 to 24°C for 5 days in a constant temperature room, the area of the disease spots was examined, the control index was found according to the evaluation standards given below, and the results in Table 2 were obtained.

Evaluation Criteria

The extent of an outbreak on the plant samples was observed visually during the examination and a 7-level control index was obtained as follows for the control effect.

(Control Index) (Extent of Outbreak)

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2. Area of disease spots less than 40 to 70% of untreated section
1. Area of disease spots 70% or more of untreated section

Table 2

| Aqueous dispersion sample | Active ingredient compound | Active ingredient concentration (ppm) | Control index |
|---|---|---|---|
| Aqueous dispersion No. 1 | Compound No. 1 | 31 | 7 |
| Comparison product of aqueous dispersion No. 1 | " | " | 2 |
| Aqueous dispersion No. 2 | Compound No. 8 | 31 | 7 |
| Comparison product of aqueous dispersion No. 2 | " | " | 1 |
| Aqueous dispersion No. 3 | Compound No. 14 | 31 | 5 |
| Comparison product of aqueous dispersion No. 3 | " | " | 1 |

Test Example 2: Test of effect in treating cucumber downy mildew

(Preparation of aqueous dispersion)

Each aqueous dispersion sample was prepared as in Test Example 1.

((Biological Test Method and Results)

Cucumber (cultivar: Suiyo) was cultivated in 7.5 cm plastic pots and inoculated by being sprayed with a suspension of *Pseudoperonopora humuli* zoospores when the seedlings reached the 2-leaf stage. After 24 hours, each aqueous dispersion sample was sprayed at a proportion of 10 mL per $0.25m^2$. After maintaining this at 22 to 24°C for 6 days in a constant temperature room, the area of the disease spots on the 1st leaf was examined, the control index was found according to the evaluation standards in aforesaid Test Example 1, and the results in Table 3 were obtained.

Table 3

| Aqueous dispersion sample | Active ingredient compound | Active ingredient concentration (ppm) | Control index |
|---|---|---|---|
| Aqueous dispersion No. 4 | Compound No. 1 | 2 | 7 |
| Comparison product of aqueous dispersion No. 4 | " | " | 3 |
| Aqueous dispersion No. 5 | Compound No. 8 | 31 | 7 |
| Comparison product of aqueous dispersion No. 5 | " | " | 2 |
| Aqueous dispersion No. 6 | Compound No. 12 | 31 | 5 |
| Comparison product of aqueous dispersion No. 6 | " | " | 1 |
| Aqueous dispersion No. 6 | Compound No. 14 | 8 | 7 |
| Comparison product of aqueous dispersion No. 6 | " | " | 1 |

(Advantages of the Invention)

According to the control method of the present invention, and particularly, the pest control effect and effect for treating plant diseases may be improved remarkably, which is effective in reducing the amount of active ingredient compound used.